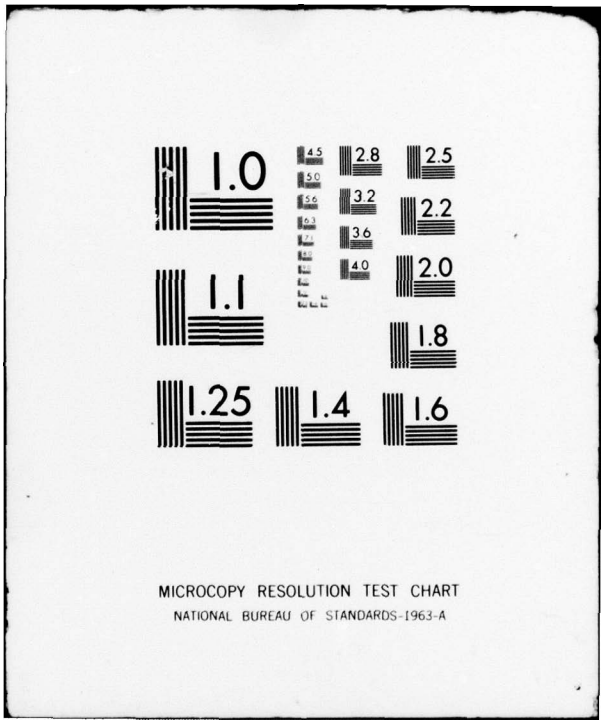
AD-A053 453 WISCONSIN UNIV-MADISON MATERIALS SCIENCE CENTER F/G 7/4
MONTE-CARLO MODELING OF PHASE CHANGES IN THE CHEMISORPTION SYST--ETC(U)
DEC 77 W Y CHING, D L HUBER, M FISHKIS N00014-76-C-0727
UNCLASSIFIED NL

| OF |
AD A053453

END
DATE FILMED
6-78
DDC


1.0
1.1
1.25
1.4
1.6
1.8
2.0
2.2
2.5
2.8
3.2
3.6
4.0
4.5
5.0
5.6
6.3
7.1
MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER: N00014-76-C-0727-3
2. GOVT ACCESSION NO.:
3. RECIPIENT'S CATALOG NUMBER:

4. TITLE (and Subtitle): MONTE-CARLO MODELING OF PHASE CHANGES IN THE CHEMISORPTION SYSTEM O/W(110).

5. TYPE OF REPORT & PERIOD COVERED: Technical Report

6. PERFORMING ORG. REPORT NUMBER:

7. AUTHOR(s): W.Y. Ching, D.L. Huber, M. Fishkis, M.G. Lagally

8. CONTRACT OR GRANT NUMBER(s): N00014-76-C-0727

9. PERFORMING ORGANIZATION NAME AND ADDRESS: Board of Regents of the University of Wisconsin System, 750 University Avenue, Madison, WI 53706

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: NR 392-014

11. CONTROLLING OFFICE NAME AND ADDRESS: Office of Naval Research, Arlington, VA 22217

12. REPORT DATE: 16 Dec 77

13. NUMBER OF PAGES: 7 (12) 9p.

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office): Office of Naval Research, Branch Office Chicago, 536 S. Clark St., Rm. 286, Chicago, IL 60605

15. SECURITY CLASS. (of this report): Unclassified

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE:

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

DDC RECEIVED JAN 24 1978 F

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

Extended Abstract, to be published in J. Vac. Sci. Technol. March/April 1978

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Surfaces, chemisorbed layers, adatom-adatom interactions, phase transitions, Monte-Carlo calculations

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Monte-Carlo calculations have been made for a number of different models of adatom interactions in the chemisorbed layer W(110)p(2x1)-O to simulate observed phase transitions. In particular the island shape is taken into account.

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE S/N 0102-014-6601

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

410 010

OFFICE OF NAVAL RESEARCH
Contract No. N00014-76-C-0727-3
Project No. NR 392-014

TECHNICAL REPORT

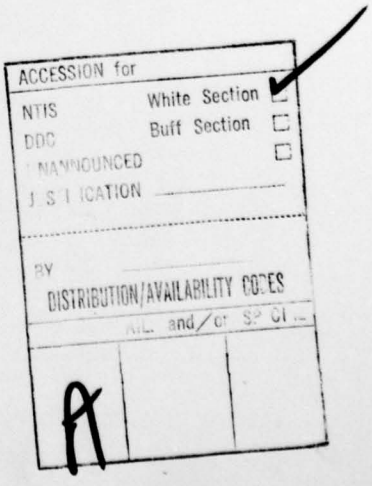
MONTE-CARLO MODELING OF PHASE CHANGES IN THE CHEMISORPTION SYSTEM O/W(110)

by

W.Y. Ching, D.L. Huber, M. Fishkis, and
M.G. Lagally
Materials Science Center
University of Wisconsin
Madison, Wisconsin 53706

December 16, 1977

To be published in J. Vac. Sci. Technol.



MONTE-CARLO MODELING OF PHASE CHANGES IN THE CHEMISORPTION SYSTEM O/W(110).*

W.Y. Ching, D.L. Huber, M. Fishkis, and M.G. Lagally†,††

Materials Science Center
University of Wisconsin, Madison 53706

Extended Abstract of Paper Presented at the AVS Symposium, Boston, Nov. 1977

* Supported by the Office of Naval Research

† H.I. Romnes Fellow

†† Please direct all correspondence to this author.

Monte-Carlo modeling of the experimentally determined transition temperature[1] as a function of coverage is reported for the chemisorption system W(110)p(2x1)-O. This system is one of the first to be studied in detail that is "closed"; i.e., it is in equilibrium neither with the gas phase nor with O dissolved into bulk W. Thus a determination of the temperature-coverage phase diagram presents the possibility of studying the thermodynamics of two-dimensional systems and through this the study of adatom-adatom interactions that lead to the formation of two-dimensional ordered phases. Additionally, of course, through use of the lever rule the concentration of adsorbed atoms in the ordered vs the disordered phase at any temperature and coverage can easily be determined.

The measurements to which the Monte Carlo calculations are compared were taken in a simple LEED diffractometer consisting of goniometer, Faraday cup collector, and electron gun. The angular distribution of intensity in a superlattice [e.g. (1/2 1/2)] reflection was measured as a function of coverage and temperature. The peak intensity can be related to the transition temperature,[1] while the angular width reflects the size of the ordered regions, or islands.[2]

The ordering of overlayer structures in general involves several interactions. Thus in modeling this ordering, a variety of experimental inputs is required. In the past, usually only the transition temperature at fixed coverage was fitted. A better approach is to fit transition temperatures as a function of coverage, but even here, a number of models with different interactions are satisfactory. An additional

experimental quantity that can be fitted is the island shape at low coverage. The present results demonstrate that the latter is a useful parameter in selecting the proper interactions.

Monte-Carlo calculations were performed for several models of the adatom-adatom interactions in the p(2x1)-O overlayer on W(110). The first corresponds to the model used in an analytical determination of the adatom-adatom interactions.[1] The others represent two additional possibilities for the interactions. All gave good agreement with the transition temperature and varying but still reasonable agreement with the decay of the superlattice beam intensity (proportional to the square of the order parameter). All, however, gave incorrect island shapes.

The experimentally observed diffraction features are round, implying that on the average the islands are round. Since only two orientations are possible by symmetry, and these do not interfere with each other, this implies that individual islands are round. Model 1 (see Fig. 1) gave islands elongated along the open direction, implying the net attractive interaction along this direction was chosen relatively too strong in the model. Model 2 gave a slight elongation in the close-packed direction, implying relatively too much net attractive interaction along this direction. Model 3[3] had a poor tendency to order at temperatures where the other two were well-ordered, and indicated row formation as in Model 2.

Figure 2 shows the calculated island size distribution at two different temperatures and a coverage of $\theta = 0.2$ for Model 2. At the

lower temperature, the adsorbed atoms are more or less in one ordered island, with an equilibrium density of adatoms in the "sea". The tendency toward elongation of the island mentioned above is evident. At the higher temperature, several smaller islands exist, with both orientations now present. The size of these islands corresponds quite well with the experimentally observed sizes.[2]

The results as listed in Figure 1 still have too wide a range of uncertainty for meaningful comparison to calculations of the indirect oscillatory interaction in chemisorbed layers.[4,5] However, in this particular system an r^{-5} decay[4,5] appears to be too rapid. An extension of the experimentally determined phase boundaries to lower coverages, as well as continued Monte-Carlo modeling to improve the fit to island shapes, is in progress.

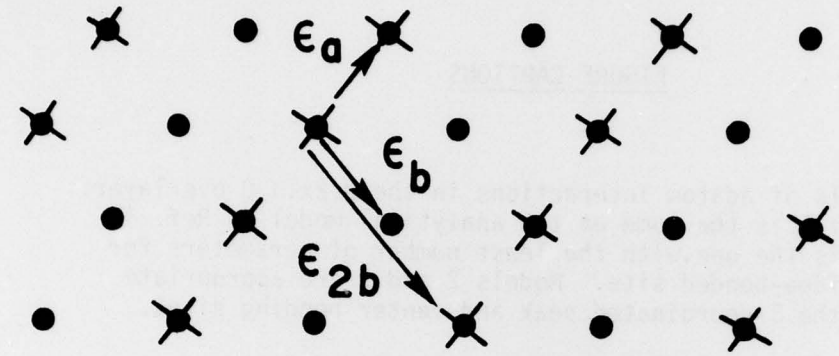
Further Monte-Carlo calculations are also underway with a goal of providing a qualitative description of the phase boundaries over the full coverage range $0 \leq \theta \leq 1$. For coverages greater than $\theta = 0.5$, the phase diagram looks quite different[1] than for $\theta < 0.5$. To fit these data requires the inclusion of three-body forces in the calculations. Detailed results of this work will be reported elsewhere.[6]

1. T.-M. Lu, G.-C. Wang, and M.G. Lagally, Phys. Rev. Letters 39, 411 (1977); G.C. Wang, T.-M. Lu, and M.G. Lagally, submitted to J. Chem. Phys.
2. G.-C. Wang and M.G. Lagally, submitted to Surface Science.
3. This model is similar to that of E. Williams, S. Cunningham, and W.H. Weinberg, this issue.
4. T.E. Einstein, CRC Critical Reviews of Solid State Sciences, to be published.
5. K.H. Lau and W. Kohn, to be published.
6. W.J. Ching and D.L. Huber, to be published.

FIGURE CAPTIONS

Figure 1 Models of adatom interactions in the p(2x1) O overlayer. Model 1 is the same as the analytical model of Ref. 1, and is the one with the least number of parameters for a bridge-bonded site. Models 2 and 3 are appropriate for the 3-coordinated peak and center bonding sites.

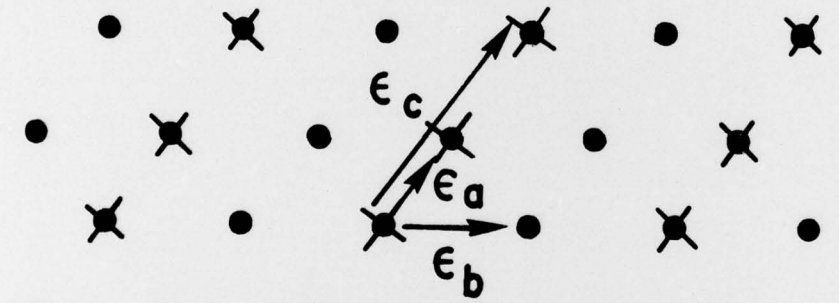
Figure 2 Monte-Carlo calculation of the distribution of adatoms on a 30 x 30 lattice for Model 2 at two different temperatures. a) 275°K, order parameter = .972, b) 325°K, order parameter = .804. Each atom has had on the average 700 chances to take a step.



MODEL 1

$\epsilon_a = \epsilon_{2b} = -0.069$ eV

$\epsilon_b = +0.15$ eV

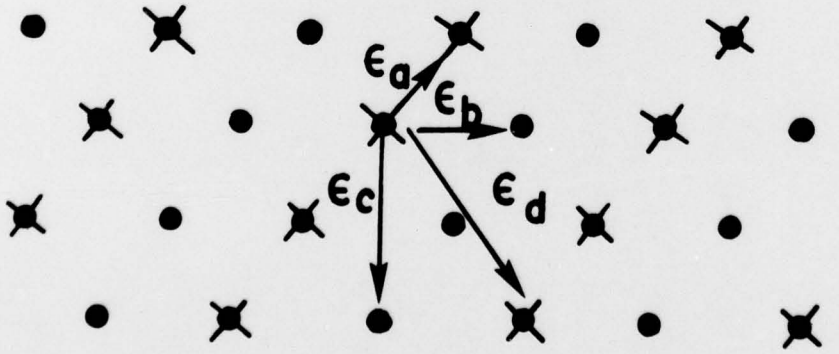


MODEL 2

$\epsilon_a = -0.072$ eV

$\epsilon_b = +0.080$ eV

$\epsilon_c = -0.049$ eV

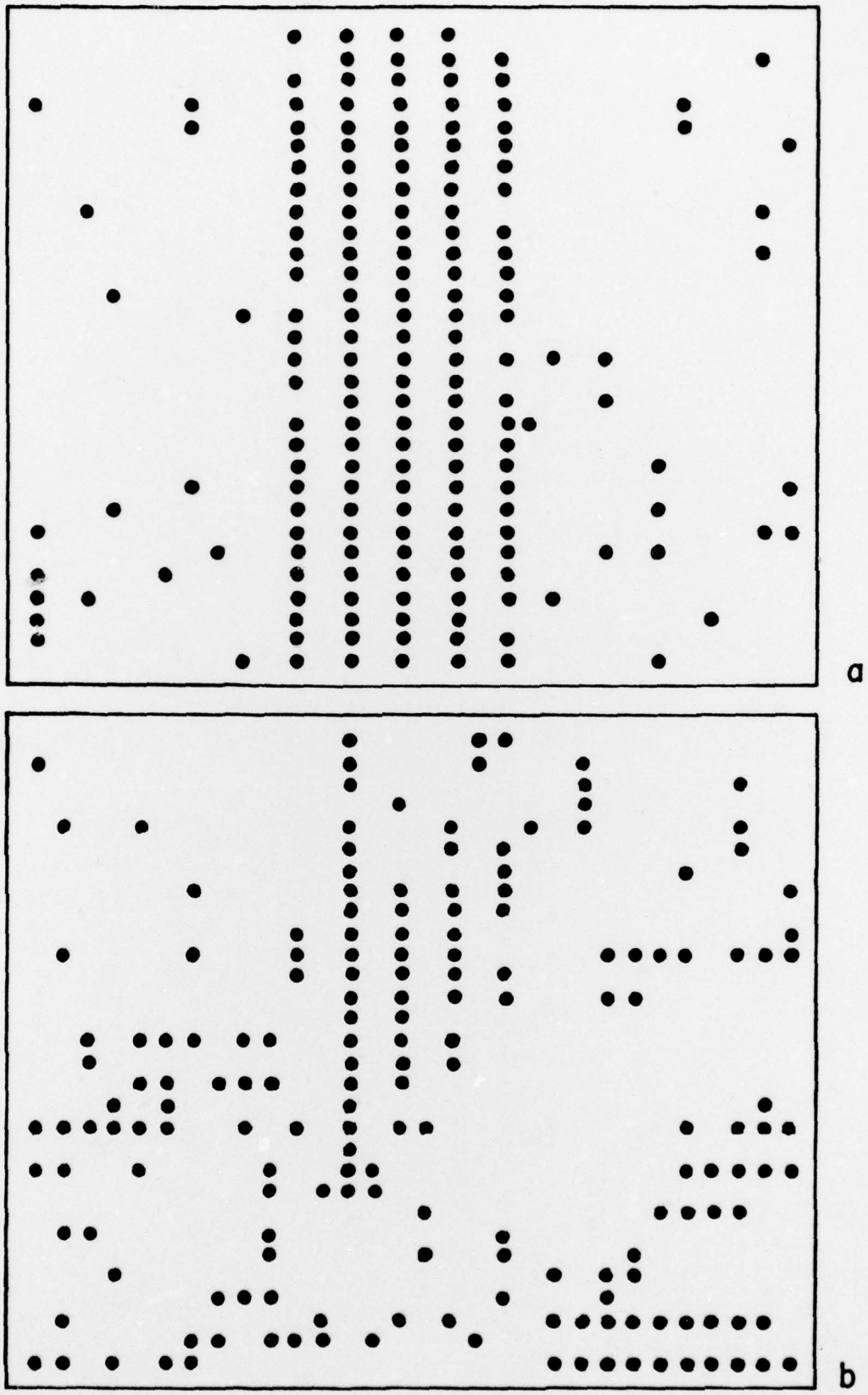


MODEL 3

$\epsilon_a = -0.09$ eV

$\epsilon_b = \epsilon_c = +0.075$ eV

$\epsilon_d = -0.03$ eV


a
b